SCI. REP. KYOTO PREF. UNIV. (NAT. SCI., LIV. SCI. & WELF. SCI.), No. 21, Ser. A, p. 17~24 (OCT. 1970)

On the Au-Fe Alloy Sample for the Perturbed Angular Correlation Experiment

By

MITSUHIRO KAWAMURA and TSUYOSHI TOMIYAMA*

(Received July 31, 1970)

The techniques for the preparation of Au-Fe alloy sample will be described in detail. It was found that the result of the activation of the sample by the Bremsstrahlung gamma rays was satisfactory for our experiment. The result of the preliminary test of the perturbed gamma-gamma angular correlation will be also mentioned.

§ 1 Introduction

The 6.18 day Au^{196} decays to Pt^{196} and Hg^{196} by the electron capture and beta emission respectively, as shown in Fig 1.[1]

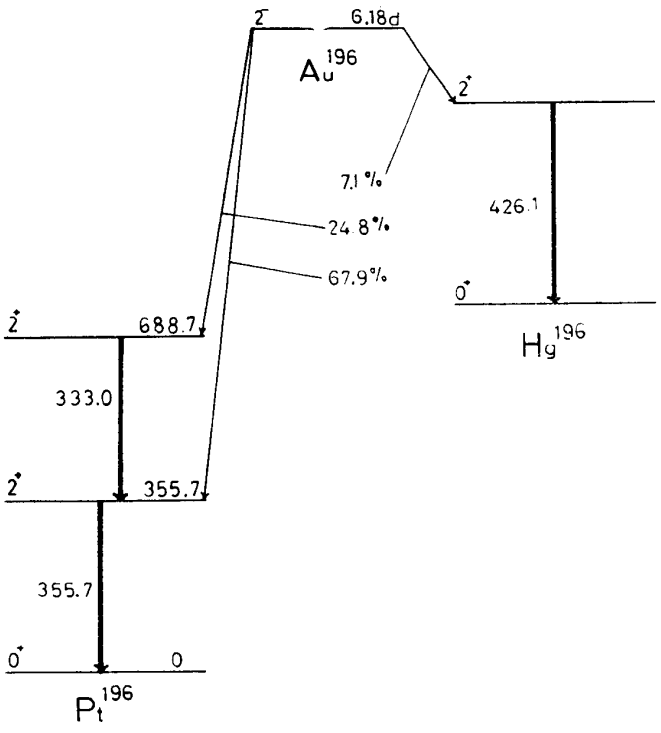

Fig 1 Decay Scheme of Au^{196}

The spins and parities of ground, first and second excited state of Pt^{196} are $0,^{+}$ 2^{+} and 2^{+} respectively. These assignments of levels and level spacings have been explained on the basis of so-called vibrational model. In order to investigate the nuclear model for even-even nucleus more precisely, however, the imformation on the nuclear moment of the excited state should be obtained. The purpose of our study is to determine the g-factor of 356 keV first excited state of Pt^{196} from the observation of the perturbation effect on the angular correlation pattern. But, it is very difficult and practically impossible to detect the effect arised from the interaction between the magnetic moment of first excited state and the external magnetic field of the order of 20~30 kgauss, because the half life[2] of the first excited state of Pt^{196} being 42 ps is too short to detect the effect. Recently, it was found from Mössbauer experiment and the other experiments that in many cases, there exist very strong internal magnetic

*Okayama College of Science

field acting on the impurity nucleus in iron alloy.[3] On the basis of this finding, the detection of the perturbation effect in gamma-gamma angular correlation has turned out to be possible by the utilization of the interaction between the nuclear moment of the excited state and the internal magnetic field. This work is based on this principle. In order to perform the experiment successfully, the following conditions in source material should be fulfilled. Firstly, the internal magnetic field on the position of Pt^{196} nucleus in alloy should be strong enough and nextly, Pt^{196} nucleus should be distributed uniformly in alloy so that every Pt^{196} nuceus could feel the internal field so strongly. This report will describe mainly the techniques of the preparation of Au-Fe alloy sample and will also mention of the preliminary test of the perturbed angular correlation experiment.

§ 2 The preparation of Au-Fe alloy samle

It is known that the order of 1390 kgauss of internal magnetic field[4] act on Pt nucleus in Pt-Fe alloy. Therefore, if iron will be alloyed with gold and part of which is produced to be radioactive Au^{196}, the daughter nucleus Pt^{196} in alloy will be expected to feel very strong internal magnetic field. In order to make up this situation, Au-Fe alloys containing Au of the order of few atomic percent were made by the following three ways.

(1) Tablets containing pure gold and pure iron powder which were both weighted so that the tablets turned out to be 2AT.% and 5 AT.% Au-Fe alloy finally, were melted by levitation furnace These alloys will be denoted as 2TL and 5Tl hereafter.

(2) Tablets mentioned above were melted by arc-melt furnace and were melted again by levitation furnace. These alloys will be denoted as 2AL and 5AL hereafter. In arc melting, vessel containing Ti metal and the tablet was evacuated down to 1×10^{-3} mmHg. After that procedure, Ti metal and tablet were melted alternately so as to absorb oxygen into Ti metal completely.

(3) Chloroauric acid ($HAuCl_4 \cdot 4H_2O$) and iron oxide (Fe_2O_3) which were both weighted so that the alloy made from these turned out to be 2AT.% and 5AT.% Au-Fe alloy finally were mixed together with distilled water and its mixture was reduced by breaching method and its product was melted by levitation furnace.

These alloys will be denoted as 2BL and 5BL hereafter. Breaching in this procedure was performed as follows. The furnace tube through which pure hydrogen gas from palladium diffuser flowed at a rate of about 7 l/hr was heated up to 1000°C and the mixture in this tube was reduced for 3 hours. The reason for the use of levitation melting in the final stage of three cases mentioned above were following one. The possible undesired intermixing of impurity to alloy will be avoided because there is no chance for sample material in hot state to contact with the vessel and furthermore, the levitation melting was expected to be best method to make uniform alloys.

The levitation melting was performed as follows. Vessel of furnace was evacuated well and filled with herium gas up to the pressure of 1 atm. Sample in the vessel was melted for 5 min. at 1800°C and cooled abruptly in copper container.

Source sample for the angular correlation measurement was shaped to be 3mm$\phi\times$3mm

and 2mmϕ×3mm cylindrical form considering the gap of the electromagnet[5] of our laboratory being 4mm.

§ 3 Activation by the Bremsstrahlung gamma rays

The samples 5AL (3mmϕ×3mm) and 5BL (2mmϕ×3mm) were irradiated by the Bremsstrahlung gamma rays from the Pt converter bombarded by KUR and JAERI electron Linac for 10 hours and 6.75 hours respectively. KUR and JAERI Linac were operated at electron energy 17MeV and 20MeV and mean current 40μA and 50μA respectively. Activities of Au^{196} produced were 30μC and 56μC for the irradiation of KUR and JAERI Linac respectively. The gamma spectrum of sample at 2 days after bombardment by KUR Linac is shown in Fig 2. The peaks at 333 keV and 356 keV correspond to the de-excitation gamma rays from first excited state to ground state and that from second to first excited state of Pt^{196} respectively and peak at 426 keV represent the de-excitation gamma rays from first excied state to ground state of Hg^{196}. These gamma rays follow from the decay of 6.18 day Au^{196}. The peak of 411 keV gamma ray is due to the decay of 2.3 day Au^{198} produced by Au^{197} (n, γ) Au^{198} reaction. There is no gamma ray of the other activity as seen in Fig 2.

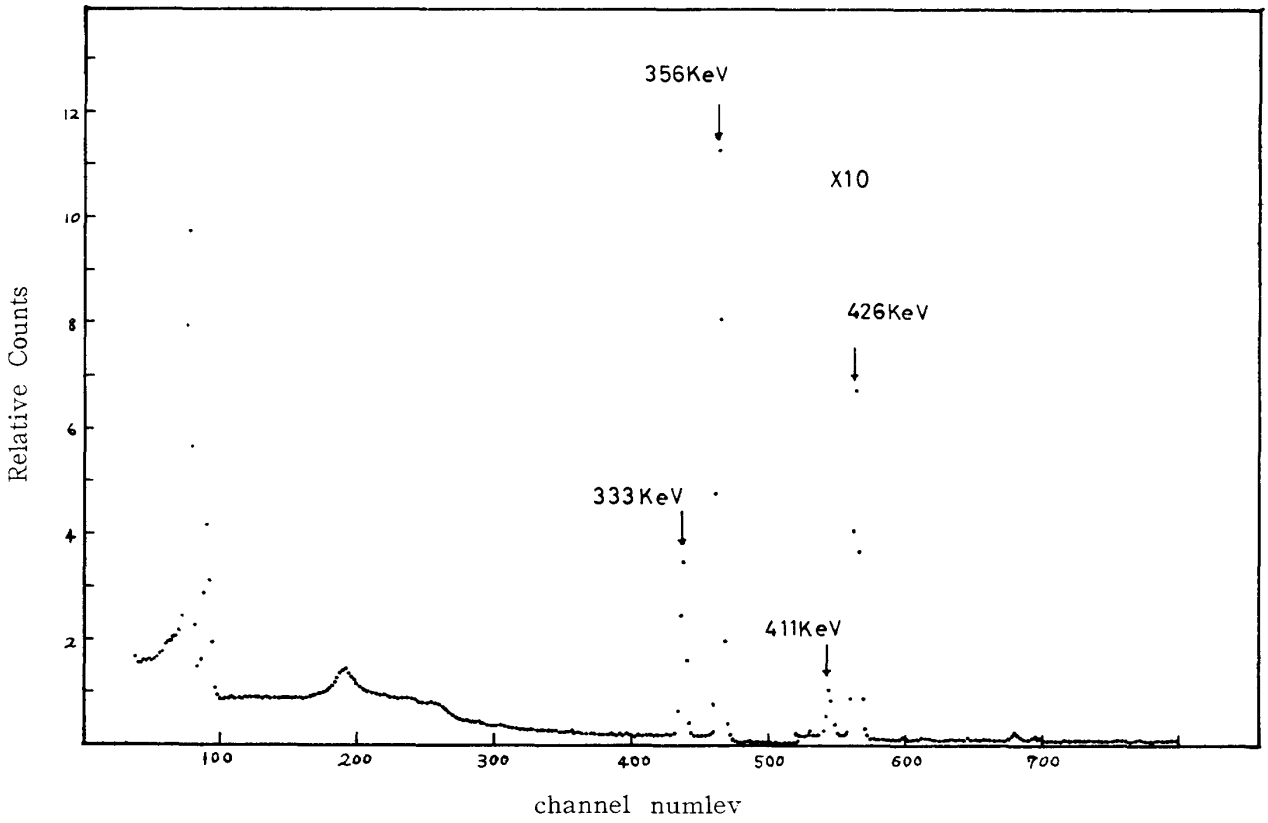

Fig 2 Gamma-ray spectrum of 5AT. % Au-Fe alloy sample irradiated by the Bremsstrahlung gamma-rays

Neutron yield from (γ, n) reaction is represented generally by the following formula.[6]

$$Y(E_o)=\int_{E_{IT}}^{E_o}\sigma(E_\gamma)P(E_\gamma,E_o)\eta(E_\gamma)dE_\gamma$$

here, $Y(E_o)$: neutron yield by (γ,n) reaction using the electron beam of its energy E_o

$\sigma(E_\gamma)$: (γn) cross section integrated over angles for the gamma rays of its energy E_γ

$P(E_\gamma,E_o)$: gamma ray spectrum of Bremsstrahlung obtained by the incident electron beam of its energy E_o

$\eta\ (E_\gamma)$: (γn) neutron detection efficiency

E_{IT} : threshold energy of (γn) reaction

For E_o close to E_{IT}, the dependence of $Y(E_o)$ to E_o and E_{IT} is represented by following formula[6].

$$Y\ (E_o) \propto (E_o-E_{IT})^m$$

In the case of the Au target,

$$Y\ (E_o) \propto (E_o-7.9)^{1.7}$$

because $E_{IT}=7.9$ and $m=1.7$[7].

The dependence of $Y\ (E_o)$ to atomic number of target nucleus will be considered to obey the general trend[8] obtained by the study group of 300MeV Linac of Tohoku University using 30MeV electron beam as shown in Fig. 3. The feature of this curve is that $Y\ (E_o)$ increases monotonically with atomic number and $Y\ (E_o)$ for Au has a value about ten times as large as for Fe. The fact that there was no harmful activity in source material is considered to reflect this trend of $Y\ (E_o)$ on atomic number and the very long or short half life of all product activity produced by Fe (γn) reaction. Judging from the experimental fact and the consideration mentioned above, it is very useful to utilize (γn) reaction to the heavy element-iron alloy, especially for the case that there is no way for production of desired activity by thermal neutron irradiation.

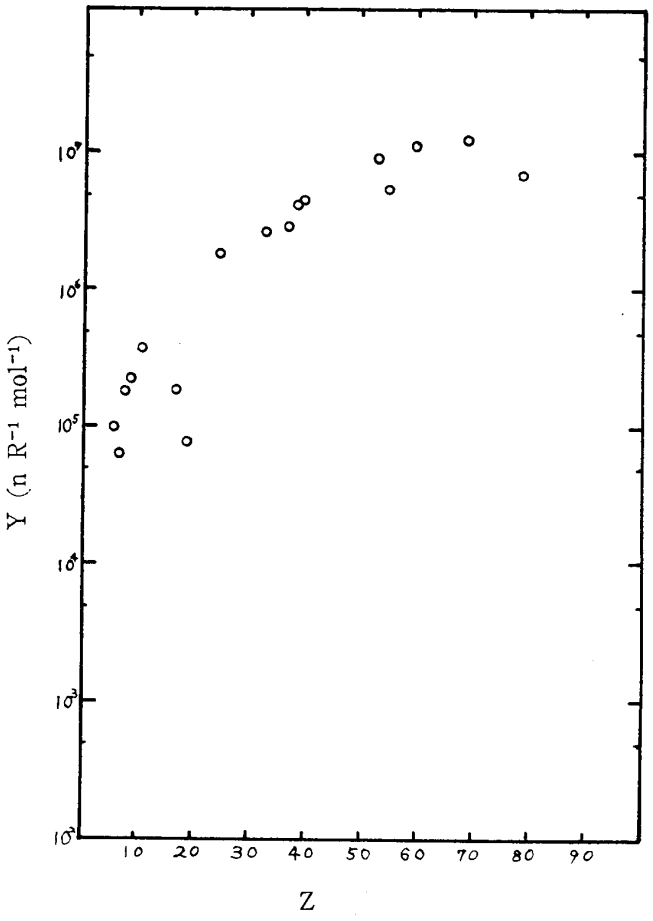

Fig 3 Neutron yield of (γ, n) reaction by 30 Mev Bremsstrahlung gamma rays as a function of atomic number

In general, activity produced by (γn) reaction due to Bremsstrahlung gamma rays is represented as follows,

$$A=Y \cdot R \cdot M\ (1-e^{-\lambda t})$$

here, A : activity produced (dpm)

Y : neutron yield of target nucleus ($n\ R^{-1}mol^{-1}$)

R : gamma ray intensity ($R\ min^{-1}$)

M : mole number of target nucleus

λ : disintegration constant of product

nucleus (min^{-1})

t : bombarding time (min)

The gross values of Y and R for KUR Linac and JAERI Linac used are shown in tablel together with the characteristic values of operation of Linacs.

Table I

Linac	KUR	JAERI
E (MeV)	17	20
Pulse width (μs)	3	4
Repetition (cps)	50	255
R (R min^{-1})	1.9×10^{6}	6.1×10^{6}
Y (n $R^{-1}mol^{-1}$)f or Au	6.2×10^{6}	1.0×10^{7}

The content of Au in Au-Fe alloy made by the three different procedure described in § 2 was checked by the activation analysis utilizing (γn) reaction and (nγ) reaction. The gamma ray for (γn) reaction was obtained from KUR Linac operated under condition mentioned above. The thermal neutron for (nγ) reaction was obtained from KUR atomic pile. The flux of thermal neutron was 1.93×10^{13} n cm^{-2} sec^{-1} and irradiation time was 1~2 min.

In both irradiation experiment, product activity of sample and standard Au foil were measured. The gamma ray spectrum was obtained from coaxial type Ge(Li) detector of active volume 23.85cc. The result are shown in table Ⅱ.

Table II

Sample	2TL	2AL	5TL	5AL	5BL
AT.%	2.28	2.12	4.85	4.81	4.56

As seens in TableⅡ, AT.% of Au in various alloys have a higher or lower value than the expected one systematically.

In activation analysis, annihilation gamma rays following positron decay of 12.8hr. Cu^{64} produced by Cu^{63} (nγ) Cu^{64} were carefully searched because very small content of Cu from the cooling copper base might mix in a alloy in course of the farely long melting on Cu plate in arc melt process.

But, the result showed that the total activity of positron emission containing Cu^{64} was less than 10^{-4} in weight.

From this result of activation analysis, it was found that it is most simple and useful way to make sample (TL) for few AT.% Au-Fe alloy.

§ 4 Preliminary test of the attenuation of angular correlation

Radioactive alloy source of 3mmϕ×3mm (30μC) or 2mmϕ×3mm (56μC) was annealed

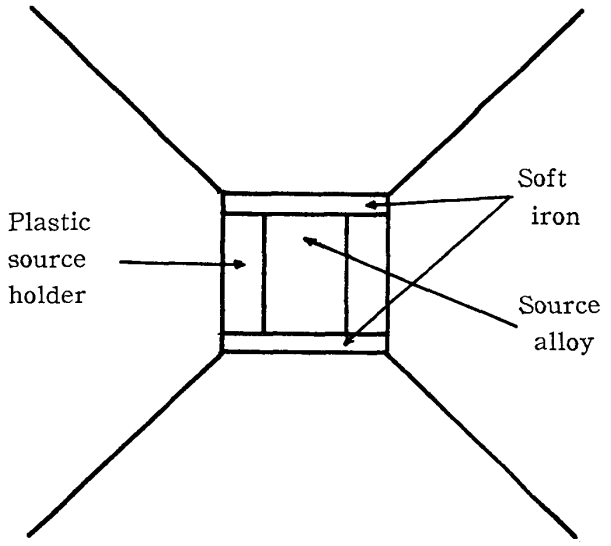

Fig 4 Setting of alloy sample in the gap of the electromagnet

in electric furnace in vacuum for 1 hour at 900°C. After that, source was placed in a gap of electromagnet as shown in Fig. 4. The gap was bridged completely by source alloy and soft iron sheet so that magnetic flux penetrate through the source alloy most effectively. The source alloy was expected to saturated at 6 kgauss of gap field which value correspond to the one in the absence of alloy.

However, alloy was saturated up to 20 kgauss of gap field because there is a report[9] that in the heavy ion implantation experiment, the direction of the microscopic magnetization in the site of impurity nucleus did not look toward that of saturation magnetization as a bulk. The gamma ray detectors for fixed and movable counter were both 1 3/4 "$\phi \times 2$" NaI (Tl) and the distance from source to crystal surface were both 12cm.

The composite peak of 333keV and 356keV in the gamma spectrum obtained by 1 3/4 "$\phi \times 2$" detector was accepted in both channel.

The change in the angular correlation pattern in applying the magnetic field was observed as an attenuation because the angular correlation pattern was an average of the shifts to the opposite directions. The measured anisotropies in the case of applying H=0 kgauss and H=20 kgauss were shown in Table Ⅲ together with the theoretical predictions.

Table III

	$A(\theta,H)=N(\theta,H)/N(90°,H)$		$\Delta A(\theta)=A(\theta,20)-A(\theta,0)$	
θ	H=0 kgauss	H=20 kgauss	Exp.	Theo.
125°	0.842±0.005	0.859±0.004	0.017±0.006	0.014
155°	1.048±0.005	1.054±0.004	0.006±0.006	0.012
180°	1.196±0.005	1.195±0.004	−0.001±0.006	0.005

The measurements were performed by using 2mm$\phi \times$3mm source alloy in order to avoid the possible effect of anisotropy due to absorption of gamma ray in source material.

The theoretical $\Delta A(\theta)$ value was obtained by the inserting the experimental values[10] of $A=+0.076\pm0.007$ and $A=+0.330\pm0.010$ and calculated values of correction factor[11] for the finite angular resolution $Q_2^2=0.9801$ and $Q_4^2=0.8281$ and the estimated[12] value of $\omega\tau=0.0785=4.5°$ into the theoretical angular correlation function derived in Appendix. Attenuation effect was detected apparently at an angle of 125°. Owing to the small effect, it is quite difficult to determine the g-factor of first excited state of Pt^{196} in the case of

the measurement of attenuation of angular correlation.

Acknowledgments

We are grateful to stuff of Takamura laboratory and Mori laboratory of Kyoto University for their help in operation of breaching apparatus and levitation furnace.

We also appreciate to the crew of KUR Linac and JAERI Linac for the operation of Linacs. We would like to thank Prof. Haseda and Prof. Endo for their helpful comment for preparation of alloys. This work was partly supported by a grant in aid for scientific research of the Ministry of Education.

Appendix

When angular correlation function for free nucleus is represented as follows,

$$W(\theta)=1+A_2P_2(\cos\theta)+A_4P_4(\cos\theta)$$

angular correlation function for the magnetic field applied up to saturation as a bulk takes the following form[13] provided that the cascade gamma rays both lies in the plane perpendicular to the direction of saturation magnetization,

$$W_S(\theta)=1+A'_2P_2(\cos\theta)+A'_4P_4(\cos\theta)$$

here, $A'_2=\bar{A}_2/\bar{A}_0$

$A'_4=\bar{A}_4/\bar{A}_0$

and $\bar{A}_0=1+1/4\ A_2+9/64A_4-(1/4A_2+5/48A_4)/(1+(2\omega\tau)^2)-7/192A_4/(1+(4\omega\tau)^2)$

$\bar{A}_2=(A_2+5/12A_4)/(1+(2\omega\tau)^2)-5/12A_4/(1+(4\omega\tau)^2)$

$\bar{A}_4=A_4/(1+(4\omega\tau)^2)$

In the non-magnetized state as a bulk, alloy is considered to consist of a many domain magnetized up to saturation at random. Consequently, angular correlation function takes a form similar to that for the polycrystalline source as follows[14],

$$W_D(\theta)=1+A_2\overline{G_{22}(\infty)}P_2(\cos\theta)+A_4\overline{G_{44}(\infty)}P_4(\cos\theta)$$

here, $\overline{G_{kk}(\infty)}=1/(2k+1)+\sum_{n\neq n'}\begin{pmatrix} I & I & k \\ n' & -n & p \end{pmatrix}^2 1/(1+(p\omega\tau)^2)$

$\omega=-B\mu/I\hbar$

$p=n-n'$.

Symbol ∞ denotes the time integrated value and B, μ, I and τ mean magnetic field, magnetic moment, spin and life time of the excited state respectively.

Especially, in the case of I=2

$$\overline{G_{22}(\infty)}=1/5+14/35\sum_{p=1}^{2}1/(1+(p\omega\tau)^2)$$

$$\overline{G_{44}(\infty)}=1/9+2/9\sum_{p=1}^{4}1/(1+(p\omega\tau)^2)$$

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